

Response to Reviewer 2

Meredith Schervish and Manabu Shiraiwa

Author comment on "Impact of phase state and non-ideal mixing on equilibration timescales of secondary organic aerosol partitioning" by Meredith Schervish and Manabu Shiraiwa, Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2022-698-AC2>, 2022

Excellent study of the interplay between thermodynamic non-ideality and mass transport limitations and how it affects partitioning and, subsequently, the characteristic equilibration time for particles.

We appreciate Reviewer 2 for the review and very positive evaluation of our manuscript. We appreciate the feedback and have responded to comments italics below.

The model is well-described and the simulations in Section 3 cover the limits that are of atmospheric interest. Minor point for this Section: The presentation in Fig. 2 is a bit confusing because it is unusual. It could be improved with the addition of some illustrations that qualitatively show the differences between the three situations.

We have added text to clarify the difference in the three simulations being run with three different activity coefficients of the condensing species in the shell phase. We have also changed the colors of the blue lines (as per Reviewer 1's suggestion) to hopefully make the plots clearer.

"Three simulations are shown where the activity coefficient is changed to represent a different miscibility of the semi-volatile species in the shell phase. The activity coefficient of the condensing species in the shell phase (γ_{shell}) is set to be (a) 1 (ideal), (b) 10^{-3} (highly miscible), and (c) 10^5 (hardly miscible), while that in the core phase is set to be 1 for all cases."

Although I believe the manuscript can be published as is, my only major recommendation concerns Section 4. It would be extremely helpful if there was a more comprehensive summary of existing measurements (even from laboratory-based surrogates) so that the reader can get a better idea of what cases tend to dominate equilibration times in the atmosphere. This would be valuable for future work. The current examples that are provide seem to focus on what may (or perhaps may not) be exceptional cases.

This is a great suggestion and we have added some context here into some of the mixing experiments done in Ye et al 2016, Ye et al 2018, and Habib and Donahue 2020. The following text has been to Section 4.

"Experimental work to probe timescales of particle-particle mixing has also been conducted by mixing two populations of particles. Ye et al (2016) showed that below 20%

RH, toluene SOA does not mix appreciably, but mixes readily with a deuterated toluene SOA population at higher RH. They also showed that even at low RH, α -pinene SOA mixes with D-toluene SOA within an hour. Ye et al (2018) extended this to include different SOA populations from isoprene, limonene, and β -caryophyllene. While the SVOC components of isoprene and α -pinene SOA mix rapidly in the presence of another SOA population, they showed that in some cases the properties of the other SOA population can inhibit this rapid mixing as in the case of toluene SOA mixing with limonene or β -caryophyllene SOA. Habib and Donahue (2022) observed mixing between erythritol-coated black carbon and sugar-coated ammonium sulfate. When using a small (and presumably less viscous) sugar under high RH and temperature conditions, the erythritol achieves a steady state in the sugar-coated ammonium sulfate population in minutes. When these conditions are changed to increase the viscosity of the particles (lower RH, lower temperature, and larger sugars), equilibrium is prolonged to a few hours. Here we discuss in detail a few cases where the results of this work may help explain some findings of Ye et al (2018). However, in this work we have only simulated one particle population and thus our interpretation is limited. Future work will focus on simulating two particle populations to represent these results."